

SOLID-STATE LASER COOLING OF YTTERBIUM-DOPED TUNGSTATE CRYSTALS

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Abstract

Anti-Stokes fluorescence can be used to cool a material if its radiative quantum efficiency is nearly unity. Until recently, the only solid which had shown net cooling was an ytterbium-doped heavy metal fluoride glass. Reasons for the lack of success with other materials include energy transfer to unwanted impurities, trapping of the fluorescence by internal reflection and reabsorption, and nonradiative relaxation via low-frequency accepting modes. However, substantial progress has recently been achieved in cooling new classes of rare-earth-doped solids. Photothermal deflection spectroscopy of Yb^{3+} in the two laser crystals $\text{KGd}(\text{WO}_4)_2$ and $\text{KY}(\text{WO}_4)_2$ demonstrates optical cooling. These materials have high figures of merit for radiation-balanced lasing, in which the Stokes heating by stimulated emission is offset by the anti-Stokes cooling due to spontaneous emission. However, approximately 1.3% of the relaxations from the excited to the ground states are nonradiative via energy transfer to impurities. In addition, about 0.3% of the pump light is absorbed per centimeter of travel by species such as transition-metal ions which dump that energy directly into heat.

Introduction

The concept of cooling a material by anti-Stokes fluorescence was proposed over seventy years ago, but it was only with the development of the laser that the idea gained experimental feasibility. Even with this tool, early failures to optically cool condensed media such as Nd^{3+} doped in yttrium aluminum garnet¹ or organic dyes dissolved in alcohol² apparently discouraged further labors. For the next couple of decades, only a smattering of papers were published, mostly restricted to theoretical analyses of thermodynamic issues; a more complete historical commentary can be found in a recent review.³

However, the field dramatically reawakened for experimentalists in the mid 1990s with the independent announcements by four different research groups of positive results. Zander and Drexhage in Germany⁴ and Clark and Rumbles in England⁵ reported net laser cooling of rhodamine dyes in ethanol; a group in Colorado achieved reduced heating of a gallium arsenide heterostructure;⁶ and investigators in New Mexico successfully cooled Yb^{3+} in a heavy metal fluoride glass.⁷ The latter work in particular has attracted attention because the medium is solid-state and consequently holds out promise as a refrigerant in certain specialized applications such as infrared satellite detectors where conventional coolers have drawbacks. Accordingly, the results on ytterbium-doped glasses have been replicated and analyzed by others, such as workers in Russia,⁸ France,⁹ Australia,¹⁰ and Spain.¹¹

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The basic scheme for solid-state laser cooling is illustrated in Fig. 1. Two bands of optically active energy levels are presumed to exist for some medium. In principle these levels could either be intrinsic to a pure material or arise by doping, quantum confinement, etc. and might for example be electronic levels of an atomic impurity, rotational-vibrational modes of a molecule, or the valence and conduction bands of a semiconductor. The levels within each band are closely spaced compared to a typical thermal energy of kT , so that Boltzmann equilibrium is always maintained, giving rise to a quasi-two-level system. The two bands will for simplicity be referred to as the ground and excited states. At a given temperature, there is some mean energy possessed by the active species in each of these bands, calculable as a centroid over the product of the thermal occupancy and the density of states; these are indicated by the dotted lines in the figure. The energy difference between these two average levels is $h\nu_F$. Now imagine pumping population from the lower to the upper manifold using laser light of photon frequency ν_P less than ν_F . On average, states near the top of the lower band get depleted while states near the bottom of the upper band get overpopulated. Assuming that intraband thermalization is rapid compared to the interband relaxation lifetime, equilibrium will be re-established in each manifold by absorbing heat in the form of phonons from the solid host. Further suppose that the fluorescence quantum efficiency of the material is very nearly unity, so that the interband relaxation is almost purely radiative, and that there are no significant transitions out of either band to other levels or species (such as by excited-state absorption or energy transfer) which decay nonradiatively. In that case, the fluorescence carries off more energy per photon (namely $h\nu_F$ on average) than the pump beam dumped into the system. Effectively, heat has been converted into light and removed from the material by anti-Stokes emission.

Last year, anti-Stokes fluorescence cooling of trivalent ytterbium doped into potassium gadolinium tungstate (KGW) was announced.^{12,13} Together with recent successes¹⁴ obtained for $\text{Yb}^{3+}:\text{YAG}$, these experiments are the first discovery of laser cooling of crystals, which have higher thermal conductivities and are easier to purify than fluoride glasses. Since Yb^{3+} in both the tungstate and garnet hosts have been demonstrated to lase, successful cooling affords the possibility of constructing a radiation-balanced laser¹⁵ which balances the Stokes heating due to the stimulated laser emission against the anti-Stokes cooling due to the spontaneous fluorescence. No external coolant is needed, nor even a separate optical cooling beam, and the pump cools the medium right where the laser emission heats it, so there is no need to conduct heat across a radius of the laser rod to its surface, from where the waste heat is usually withdrawn.

In the present paper, further details of the $\text{Yb}^{3+}:\text{KGW}$ cooling experiments are presented. The general equations for analyzing the absorption and emission spectra of a biaxial material are summarized and applied. The cooling efficiency data is presented in two different manners—an absolute efficiency in

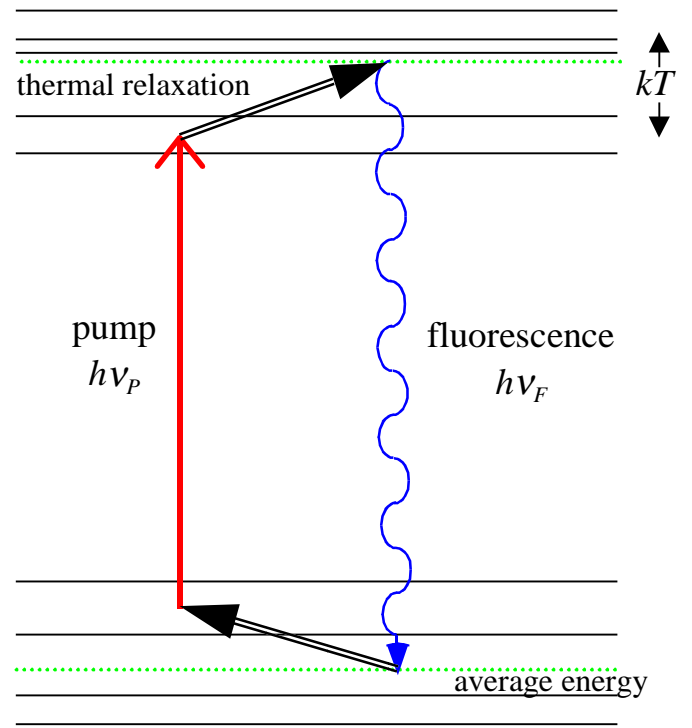


Fig. 1. Prototypical solid-state laser cooling scheme.

terms of the *incident* laser power and a relative efficiency in terms of the *absorbed* laser radiation—for all three crystallographic orientations. In addition, successful cooling of a similar material, namely ytterbium-doped potassium yttrium tungstate (KYW), is found whose figures-of-merit for radiation-balanced lasing¹³ are the highest of any known medium. A head-to-head comparison of the best cooling results for this sample and that of the fluoride glass, scaled to identical dopant concentration, host thermal conductivity, and laser pump power, shows that KYW currently achieves about six times greater cooling. This difference can only be expected to grow when cleaner tungstate samples are grown, if the history of the improvements in the fluoride glass cooling efficiencies is any indication.

Absorption and Emission Spectra

The material used in this study was KGd(WO₄)₂ doped with nominally 5 at.% Yb³⁺ in the melt which substitutes for the gadolinium. The crystals were grown commercially at their 1075 °C melting temperature using a modified Czochralski technique. The actual concentration was determined to be 3.5 at.%, corresponding to $N = 2.2 \times 10^{20} \text{ cm}^{-3}$, using the peak *a*-axis absorption cross section, $\sigma_{ab}^a(981.2 \text{ nm}) = 2.2 \times 10^{20} \text{ cm}^2$, known from an x-ray fluorescence analysis.¹⁶⁻¹⁹ The crystal structure is monoclinic and the room-temperature lattice parameters are $a = 8.095 \text{ Å}$, $b = 10.42 \text{ Å}$, and $c = 7.588 \text{ Å}$ with a 94.43° angle between the *a* and *b* axes and an average index of refraction of $n = 2.019$ varying by $4 \times 10^{-7} / ^\circ\text{C}$. The lifetime of the ytterbium ions in this host is $\tau = 0.6 \text{ ms}$ and the average thermal conductivity is $3.3 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. The RT energy-level structure of Yb:KGW consists of 4 levels (at 0, 163, 385, and 585 cm⁻¹) in the ²F_{7/2} ground state and 3 levels (at 10188, 10471, and 10682 cm⁻¹) in the ²F_{5/2} excited state.

Figure 2 shows the polarized absorption and emission cross sections of Yb:KGW near 1 μm. Even at room temperature, three peaks in absorption starting from the dominant 0–0 peak and proceeding to higher energies, and four peaks in emission running from the 0–0 peak to lower energies are clearly resolved; only the Kramers degeneracies of the levels in the ground and excited states remain. The wavelength λ_F corresponding to the mean fluorescence photon energy can be computed for a biaxial material as

$$\lambda_F = \left[\sum_{\gamma} \int \lambda I_{\gamma}(\lambda) d\lambda \right] \left[\sum_{\gamma} \int I_{\gamma}(\lambda) d\lambda \right]^{-1} \quad (1)$$

where the sum runs over the three crystallographic axes, $\gamma = \{a, b, c\}$. This gives $\lambda_F = 992.5 \text{ nm}$ for Yb:KGW, as indicated by the dashed vertical lines in Fig. 2. Here $I_{\gamma}(\lambda)$ is the spectral fluorescence intensity (in $\text{W} \cdot \text{m}^{-2} \cdot \text{nm}^{-1}$) and is proportional to the corresponding emission cross section $\sigma_{em}^{\gamma}(\lambda)$ in the following manner. The Füchtbauer-Ladenburg equation is

$$\sigma_{em}^{\gamma}(\nu) = \left(\frac{c}{n\nu} \right)^2 \frac{g_{\gamma}(\nu)}{8\pi\tau} \Rightarrow \sigma_{em}^{\gamma}(\lambda) = \frac{\lambda^4 g_{\gamma}(\lambda)}{8\pi n^2 c \tau} \quad (2)$$

where the normalized lineshape function is

$$g_{\gamma}(\nu) = [I_{\gamma}(\nu)/h\nu] \left[\frac{1}{3} \sum_{\gamma} \int d\nu I_{\gamma}(\nu)/h\nu \right]^{-1} \Rightarrow g_{\gamma}(\lambda) = 3\lambda I_{\gamma}(\lambda) \left[\sum_{\gamma} \int \lambda I_{\gamma}(\lambda) d\lambda \right]^{-1}. \quad (3)$$

Note that $I_{\gamma}(\nu)/h\nu$ is simply the spectral photon flux density (i.e., the number of emitted photons per unit time per unit area per unit frequency interval). Also note that an absolute measurement of the fluorescence intensity (which depends on the collection solid angle, detector efficiency, sample geometry, etc.) is not

needed, provided that relative values for different wavelengths and polarizations are in the correct ratio to one another. (That is, care must be taken to avoid saturation of the pump beam or reabsorption of the fluorescence, and the instrumental response must be flattened using a standard lamp.) In practice, equations (2) and (3) are used to determine the emission cross sections from the fluorescence spectra.

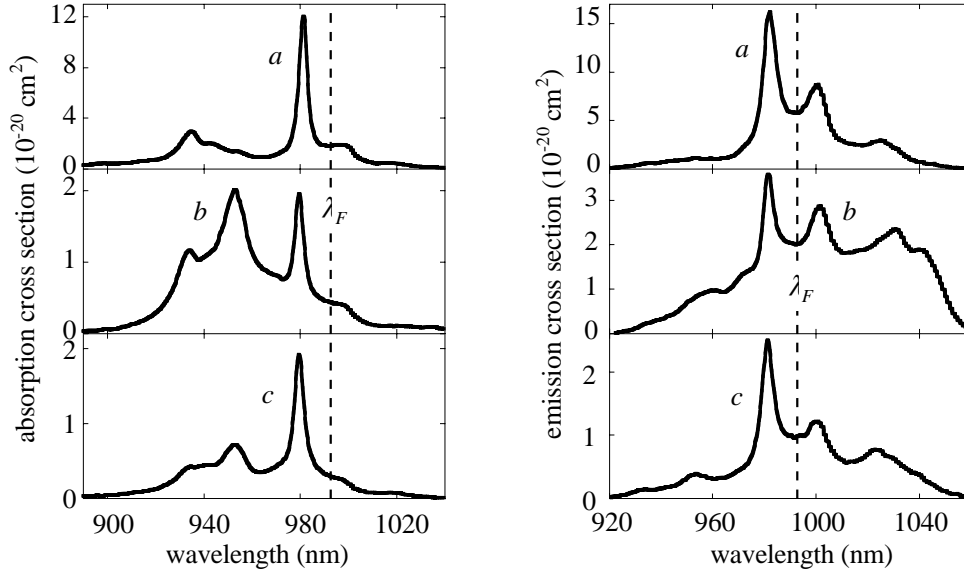


Fig. 2. Polarized optical spectra of Yb:KGW at RT.

The measured absorption coefficients, $\alpha_\gamma(\lambda)$ in cm^{-1} , can be converted to absorption cross sections by dividing them by the Yb^{3+} concentration, N . The principle of reciprocity then affords an independent check on Eq. (2),

$$\sigma_{em}^\gamma(\lambda) = \sigma_{ab}^\gamma(\lambda) \frac{Z_g}{Z_e} \exp\left[\frac{hc}{kT}(\lambda_{00}^{-1} - \lambda^{-1})\right] \quad (4)$$

where the 0–0 wavenumber is $\lambda_{00}^{-1} = 10188 \text{ cm}^{-1}$. Here the ratio of the partition functions of the ground and excited states is

$$\frac{Z_g}{Z_e} = \left\{ \sum_j \exp\left[\frac{-hc}{kT\lambda_j}\right] \right\} \left\{ \sum_i \exp\left[\frac{hc}{kT}(\lambda_{00}^{-1} - \lambda_i^{-1})\right] \right\}^{-1} \quad (5)$$

where j runs over the 4 levels in the lower manifold and i over the 3 levels in the upper band. This ratio is equal to 1.25 for Yb:KGW at room temperature. Calculating the stimulated emission cross sections via Eq. (4) gives fair agreement with the fluorescence spectra in Fig. 2.

Photothermal Deflection Spectra

As one can see in Fig. 2, substantial absorption strength remains at wavelengths longer than λ_F . Photons in this region have energies less than the mean energy of the emitted photons. Thus, according to the scheme in Fig. 1, one would expect the material to cool when pumped at such wavelengths, provided that there are no nonradiatively decaying impurities which get excited either directly by the pump laser or indirectly via energy transfer from the active ytterbium ions. (One can assume that ytterbium in the otherwise pure host has a negligible nonradiative decay rate, because there are no higher excited states to

enable excited-state absorption, and because multiphonon decay between the manifolds is suppressed by the high order of that process. This is true despite cooperative emission of pairs of Yb^{3+} ions at 3.5% concentration, which gives rise to a noticeable bluish fluorescence.²⁰⁾

An ideal absolute cooling efficiency per unit length can be defined as the ratio of the cooling power to the pump laser power,

$$\frac{1}{P_{\text{pump}}} \frac{dP_{\text{cool}}^{\text{ideal}}}{dL} = \alpha_{\gamma}(\lambda) \frac{\lambda - \lambda_F}{\lambda_F}. \quad (6)$$

This is valid provided that the pumped length of the sample is short compared to the penetration depth α_{γ}^{-1} . Equation (6) is plotted in Fig. 3 by calculating the absorption coefficients from the emission cross sections in Fig. 2 using Eq. (4). The peak value is 0.048/cm and occurs for *a*-polarization and a wavelength of 1001.2 nm; the significance of this is that if 1 cm of the sample were pumped with 1 W at this wavelength and orientation, the cooling power would ideally be 48 mW.

To test for and quantify the actual cooling, photothermal deflection measurements are performed using the “mirage” effect.^{12,13} A pump beam which can be tuned across the 1- μm absorption band of Fig. 2 is slightly displaced laterally from a counter-propagating probe beam at whose wavelength the sample is transparent. Specifically, a stable helium-neon laser running on its red line is used for the probe and a cw titanium-sapphire laser for the pump. At short wavelengths, the volume of the sample in the vicinity of the pump beam warms up with a time constant of a few dozen milliseconds, depending on the geometry and the thermal constants of KGW. Consequently there is a radial temperature distribution and hence a gradient in the refractive index proceeding outward from the center of the pump beam. The probe beam is positioned near the steepest part of this gradient and is refracted toward the center of the pump beam because dn/dT is positive. That is, the HeNe beam exits the sample at an angle to its original direction of propagation. After traveling a substantial distance in air, the probe beam is therefore displaced in comparison with its position when the pump laser is periodically switched off using a shutter. This deflection is synchronously measured using a position-sensitive detector. The peak deflection amplitude is proportional to the temperature change and hence to the net energy deposited in the sample. By tuning the pump laser through the mean fluorescence wavelength λ_F , this amplitude diminishes to zero and then the direction of the deflection changes sign and regrows in amplitude as the sample begins to laser cool. Ratioing out the sample absorptance and laser power thus gives a measurement of the actual relative cooling efficiency, defined as the ratio of the cooling to the absorbed pump power. This can be written as³

$$\frac{P_{\text{cool}}^{\text{actual}}}{P_{\text{abs}}} = \frac{\eta\lambda - \lambda_F}{\lambda_F} - \frac{\alpha_{\gamma}^{\text{impurity}}(\lambda)}{\alpha_{\gamma}(\lambda)}. \quad (7)$$

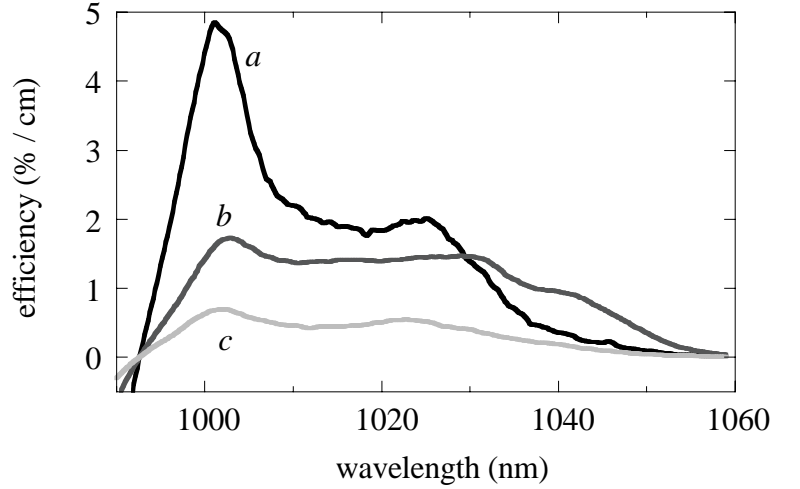


Fig. 3. Percent absolute cooling efficiency per unit length in the absence of impurity heating.

Here η is the quantum efficiency of the relaxation from the ytterbium excited state back down to the ground state; a value lower than unity represents a net heating rate due to energy transfer to quenching centers as a fraction of the Yb^{3+} emitted power in the absence of energy transfer. The cooling efficiency can also be reduced by direct absorption of the pump laser by “dark impurities” such as transition metals which are not involved in the ytterbium excitation-relaxation cycles at all and hence do not affect η . These can be modeled by a polarization-dependent absorption coefficient $\alpha_{\gamma}^{\text{impurity}}$ which is presumably broadband and hence only depends weakly on the pump wavelength λ .

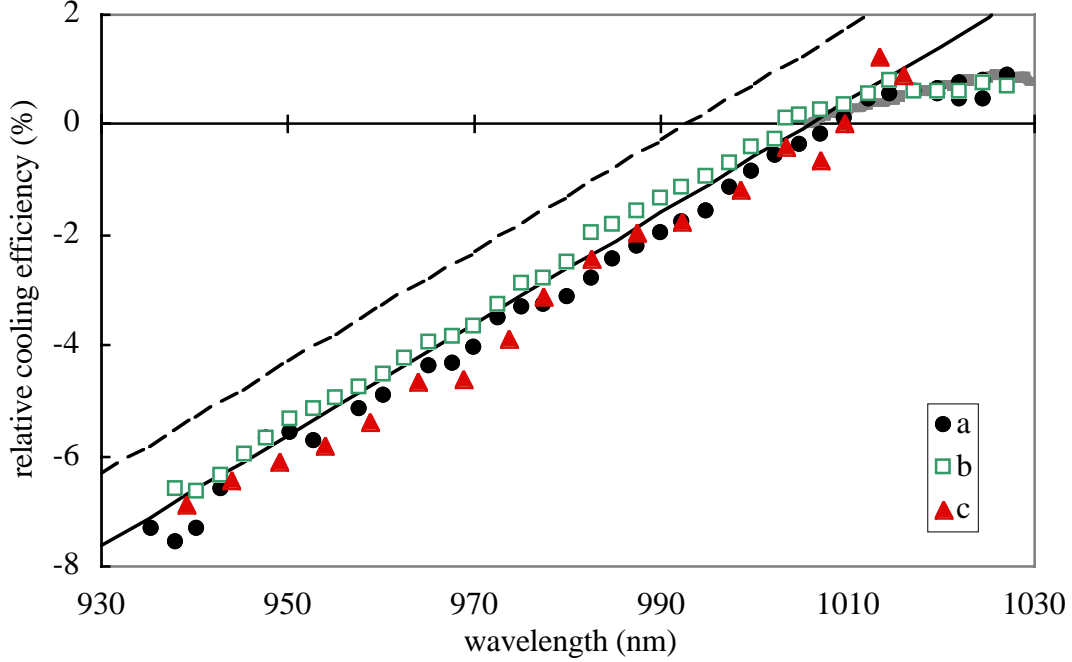


Fig. 4. Percent relative cooling efficiency of Yb:KGW and model fits.

The photothermal data, normalized to the laser power and Yb^{3+} absorptance, is plotted in Fig. 4. The dashed line is the ideal relative cooling efficiency obtained for unit quantum efficiency and zero impurity absorption; the experimental data have been scaled to the same slope. The straight solid line is the cooling efficiency obtained when the the ytterbium fluorescence quantum efficiency is reduced to $\eta = 0.987$. The fact that all three sample polarizations give curves which essentially follow this line suggests that energy transfer to quenching impurities is the dominant heating mechanism, as it would be very surprising if $\alpha_{\gamma}^{\text{impurity}}$ just happened to give the same 1.3% reduction in efficiency for all three orientations. However, there remains a small polarization-dependent residual on the heating side, as well as a significant drop-off in the cooling at the longest wavelengths. Both of these imply a small amount of direct absorption of the pump light by nonradiatively relaxing impurities. Concentrating on the long-wavelength cooling region alone, for example, the thick grey curve in Fig. 4 is a plot of the effect of setting $\alpha_b^{\text{impurity}} = 0.003 \text{ cm}^{-1}$. This absorption coefficient is so small that there is no way to directly detect it in the b -axis absorption spectrum of Fig. 2. But because every photon these dark impurities absorb gets converted entirely into heat, it nevertheless has a substantial effect in the cooling region. This is what makes laser cooling of solids so challenging from a materials point of view. Nevertheless the results in Fig. 4 are encouraging of further work because no special effort has yet been made to purify the commercially obtained samples used here.

While plotting the relative cooling efficiency as in Fig. 4 is a good way to probe for impurity effects, it does not permit a comparison to be made of the potential performance of different materials. After all, the ideal relative efficiency is $(\lambda - \lambda_F) / \lambda_F$ and $\lambda_F \approx 1 \mu\text{m}$ for Yb^{3+} , so such a plot looks essentially identical for any ytterbium-doped cooler. One would like to compare experimental temperature drops for samples scaled to the same concentration, dimensions, thermal conductivity, and pump power. Such a comparison is made in Fig. 5 for two hosts: potassium yttrium tungstate (KYW), a material chemically similar to KGW but with a slightly higher cooling figure-of-merit,¹³ and zirconium-barium-lanthanum-aluminum-sodium fluoride glass (ZBLAN). This figure shows that the tungstate host can achieve about 6 times greater cooling than the heavy metal fluoride glass under the same conditions.

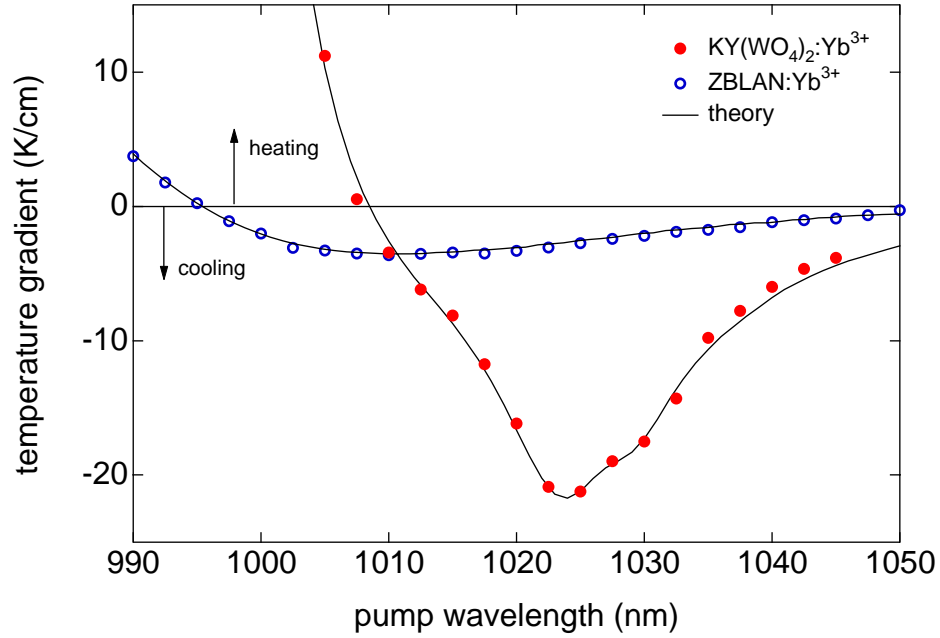


Fig. 5. Comparison of the cooling performance of Yb^{3+} doped in KYW and ZBLAN.

Summary

Laser cooling of ytterbium-doped tungstate crystals has been demonstrated and quantified by photothermal deflection spectroscopy. The commercially obtained samples have a fluorescence quantum efficiency of 98.7% and a dark impurity absorption coefficient of 0.003 cm^{-1} , indicating that further purification will be required before these materials will be ready for practical applications. Nevertheless, they already show promise of surpassing the performance of Yb^{3+} in the only other hosts in which cooling has been demonstrated, namely the heavy metal fluoride glass ZBLAN and yttrium aluminum garnet. Favorable properties of the ytterbium-tungstates include exceptionally high absorption and emission cross sections, large thermal conductivity, the fact that they can be doped at up to 10 at.%, and substantially lower impurity heating than good commercial sources¹² of YAG or YLF. In addition, their high lasing slope efficiency (up to 87%)¹⁸ and figures-of-merit more than an order of magnitude larger¹³ than those of either ZBLAN or YAG imply that the tungstates are good host candidates for radiation-balanced lasers.

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